AFOSR - TR - 72 - 1782

AFOSR-72-2332

AD 748943

UTEC CE 72-145

ON THE GENERAL THEORY OF STEKLOV - AGING MATERIALS

By

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August 1972

For presentation at the

VI^{e} Congrès International

de Rhéologie - Lyon

4 - 8 Septembre 1972

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35

UNCLASSIFIED

Security Classification

DOCUMENT CONTROL DATA - R & D

(Security classification of title, body of abstract and indexing annotation must be entered when the overall report is classified)

1. ORIGINATING ACTIVITY (Corporate author)

UNIVERSITY OF UTAH
COLLEGE OF ENGINEERING
SALT LAKE CITY, UTAH 84112

2a. REPORT SECURITY CLASSIFICATION

UNCLASSIFIED

2b. GROUP

3. REPORT TITLE

ON THE GENERAL THEORY OF STEKLOV - AGING MATERIALS

4. DESCRIPTIVE NOTES (Type of report and inclusive dates)

Scientific Interim

5. AUTHOR(S) (First name, middle initial, last name)

J EDMUND FITZGERALD

6. REPORT DATE

Aug 1972

7a. TOTAL NO. OF PAGES

32

7b. NO. OF REFS

11

8a. CONTRACT OR GRANT NO.

AFOSR-72-2332

b. PROJECT NO.

7921-02

c.

61102F

d.

681308

9a. ORIGINATOR'S REPORT NUMBER(S)

UTEC CE 72-145

9b. OTHER REPORT NO(S) (Any other numbers that may be assigned this report)

AFOSR - TR - 72 - 1782

10. DISTRIBUTION STATEMENT

Approved for public release; distribution unlimited

11. SUPPLEMENTARY NOTES

TECH, OTHER

12. SPONSORING MILITARY ACTIVITY

AF Office of Scientific Research (NAE)
1400 Wilson Boulevard
Arlington, Virginia 22209

13. ABSTRACT

A broad class of materials possessing both instantaneous nonlinear elasticity and dissipation in addition to fading memory with aging effects is described. The measure of the generalized imput function, which is a multiplet in terms of the deformation gradient, the temperature, the gradient of the temperature, as well as various chemical affinities, is given a semi-norm over a Banach Space. The semi-norm includes a summand which is a modification of the Steklov Average to P - integrable Lebesgue functions. It is assumed that the generalized response is a nonlinear function of the present input and a material property-history kernel determined by a Steklov-Lebesgue norm. Examples applied to solid propellant are given.

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DD FORM 1 NOV 65 1473

UNCLASSIFIED

Security Classification

UNCLASSIFIED

Security Classification

14 KEY WORDS	LINK A		LINK B		LINK C	
	ROLE	WT	ROLE	WT	ROLE	WT
STEKLOV AGING						
PROPELLANT AGING						
SOLID PROPELLANT						
STEKLOV-LEBESGUE NORM						
VISCOELASTICITY						

"ON THE GENERAL THEORY OF STEKLOV-AGING MATERIALS"

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SUMMARY

A broad class of materials possessing both instantaneous nonlinear elasticity and dissipation in addition to fading memory with aging effects is described. The measure of the generalized input function, Λ, which is a multi-plet in F, the deformation gradient; θ, the temperature; $g = \mathrm{grad}\theta$, as well as various chemical affinities, A_k; is given by a semi-norm over a Banach Space. With the definition of the history $\Lambda^t = \Lambda^t(s) \equiv (t-s)$; $s\varepsilon[0,\infty)$ and with the restriction of Λ^t to past history given by $\Lambda_r^t = \Lambda_r^t(s) = \Lambda_r^t(t-s)$; $s\varepsilon(0,\infty)$ the semi-norm is:

$$||\Lambda^t(s)||_{S_p^N} = \sum_{i=0}^{N}|\Lambda^{(i)}(o)|h_i(o) + \sum_{P=1}^{\infty}\left[\frac{1}{1+k\ \mathrm{measD}}\int_D|\Lambda_r^t(s)|^p h_p^p(s)ds\right]^{1/p}$$

$$\text{with } \left[\frac{1}{1+k\ \mathrm{measD}}\int_D h_p^P(s)ds\right]^{1/P} < \infty \text{ and } h_p^P(s) \geq 0 \quad . \tag{1}$$

and $\Lambda^{(i)}(0)$ the present i^{th} time rate of change.

The second summand is a modification of the *Steklov Average* [Kantorovich, 1964] to P - integrable Lebesgue functions.

It is assumed that the generalized response $\Omega(t)$ is a nonlinear function(al) of the present input $\Lambda(t)$ and a material property-history kernel determined by the *Steklov-Lebesgue* norm $||\Lambda^t(s)||_{S_p^N}$ such that

$$\Omega(t) = F\left[||\Lambda^t(s)||_{S_P^N}\right]\Lambda(t) \tag{2}$$

Equation (1) shows that as time increases mesD increases and that the influence of the past history on present response decreases. *For a given finite duration input, its influence decreases both the longer in the past it occurred and the older the material is.* This latter effect is termed *Steklov-aging*. As the age of the material becomes very large the past history effects are obliterated and from Eq. (1)

$$\lim_{\substack{t\to\infty \\ \mathrm{mesD}\to\infty}}||\Lambda^t(s)||_{S_p^N} = \sum_{i=0}^{N}|\Lambda^{(i)}(o)|h_i(o) \quad . \tag{3}$$

Thus a *Steklov-aging material under long term aging approaches the behavior of a nonlinear viscoelastic or Markoffian type material.* For very small inputs, $\Lambda(o)$ and small time rates of change of inputs, $\Lambda^{(i)}(0)$, the material, after long term aging, becomes linearly viscoelastic.

Examples of the theory as applied to solid propellants and a sand-asphalt concrete are given.

1.0 INTRODUCTION

The application of the linear and nonlinear theories of viscoelasticity has had wide application in the characterization and stress analysis of solid propellants, highly solids loaded polymers, and asphalt pavement during the past two decades.

So long as the characterization and verification of the selected constitutive equation was confined to the usual loading or straining conditions such as a constant strain rate, ramp relaxation, or constant stress creep the comparison of theory with experiment was generally satisfactory. This result was to be expected since the theory and experiment are often a curve fitting exercise over a narrow range of load types.

When viscoelastic theory is applied to the prediction of repeated loads such as a sawtooth input or multiple ramp input, the predictive results of the theory are often quite unsatisfactory. The above statement applies to linear viscoelasticity as well as to the several forms of nonlinear viscoelasticity utilizing the multiple integral approach.

Details of the previous statements are presented in [Farris and Fitzgerald, 1970], [Fitzgerald and Farris, 1970], and Chapter XI of [Fitzgerald and Hufferd, 1971]. A most excellent comparative review is given in [Stafford, 1969].

Figure 1 demonstrates the above problem based upon some of Farris' experiments, using a filled polyurethane propellant.

The use of a repeated sawtooth strain history is typified by the curves of Figure 2 for a highly solids loaded polybutadiene acrilonitrile propellant [Bennett, 1971].

Previous publications by the author and his co-workers employed the use of homogeneous functions of the strain history made specific through Lebesgue norms. The Lebesgue norm, $||\varepsilon||_{L_p}$ is essentially the weighted P-summable integral of the infinitesimal strain history, ε wherein

$$||\varepsilon||_{L_p} \hat{=} \left[\int_{\text{meas } S} |\varepsilon|^{P} h_p(s)ds \right]^{1/p} \tag{4}$$

The term $h_p(s)$ is a positive decreasing function of s for fading memory theories where P is taken as unity. Certain boundedness restrictions apply to $h_p(s)$.

For $P \to \infty$, the Chebyshev norm results wherein

$$||\varepsilon||_{L_\infty} = \text{ess. sup } |\varepsilon| \tag{5}$$

Using a rather simple expression [Farris, 1970] for stress, T, versus strain ε, wherein

$$T_{11}(t) = 435 \left[\frac{||\varepsilon_{11}||_\infty}{||\varepsilon_{11}||_{21}} \right]^{2.25} \varepsilon_{11}(t) \tag{6}$$

the results of Figure 3 were obtained [Farris, 1970].

It should be noted that the general form of Eq. (6), which is the L_∞ norm divided by the L_p norm to the n^{th} power (with $P \geq 1$), when multiplied by ε_{11} yields the following results:

•For a constant strain rate test, $\varepsilon_{11} = Rt$

$$||\varepsilon_{11}||_\infty = Rt; \quad ||\varepsilon_{11}||_P = Rt - \frac{t^{1/P}}{(P+1)^{n/P}}$$

$$T_{11} = A(P+1)^{n/P} \; Rt^{1-n/P}$$

$$\text{or} \quad T_{11} = A(P+1)^{n/P} \; \varepsilon_{11} t^{-n/P} \tag{7}$$

•For a step relaxation test $\varepsilon_{11} = \varepsilon_0$

$$||\varepsilon_{11}||_\infty = \varepsilon_0; \quad ||\varepsilon_{11}||_P - \varepsilon_0 \; t^{1/P}$$

$$T_{11} = A\varepsilon_0 \; t^{-n/P} \tag{8}$$

The last result predicts an inverse power law for the relaxation modulus, $A\ t^{-n/P}$, so that the value of A and the slope, n/P, can be determined from a step relaxation test since $G(t)_{relax.} = A\ t^{-n/P}$.

The constant strain rate test yields the secant modulus

$$G(t)_{secant} = (P+1)^{n/P} \; G(t)_{relax.} \tag{9}$$

from which P+1, hence P and n may be determined.

The predictions of Figure 3 were made using data obtained as described. A comparison of the above norm expression with linear viscoelasticity for a polyurethane propellant is given in Figure 4 [Farris, 1970].

A more general expansion of Farris' earlier expressions has been given by [Vakily and Fitzgerald, 1972] wherein the stress function

involves a sum of Lebesgue norm ratios and the naturally occurring inverse, power law kernel in the viscoelastic integral as follows:

$$T_{11}(t) = \sum_{i=0}^{N} \sum_{j=0}^{N} A_{ij} \left(\frac{||f||_{q_i}}{||f||_{p_i}} \right)^{n_i} \int_0^t (t-\tau)^{-m_j} \dot{\varepsilon}_{11}(\tau) d\tau \quad . \tag{10}$$

The first term of the above expansion, $i = j = 0$ with $q_0 = \infty$ yields

$$T_{11}(t) = A_{00} \left[\frac{||\varepsilon_{11}(t)||_\infty}{||\varepsilon_{11}(t)||_{p_0}} \right]^{n_0} \varepsilon_{11}(t) \tag{11}$$

which is the previous Farris expression, Eq. (6).

Including the current value of the strain, $|\varepsilon_{11}|$ and simplifying a three term expansion of Eq. (10) results in

$$T_{11}(t) = A_1 \left[\frac{|\varepsilon_{11}|}{||\varepsilon_{11}||_{p_0}} \right]^{n_0} \varepsilon_{11}(t) + A_2 \left[1 - \frac{|\varepsilon_{11}|}{||\varepsilon_{11}||_\infty} \right]^{n_1}$$

$$\int_0^t (t-\tau)^{-m_1} \dot{\varepsilon}_{11}(\tau) d\tau \; . \tag{12}$$

where we have defined $||\varepsilon||_0 = |\varepsilon|$.

Various other specific forms of the above Lebesgue norms may be formulated.

Applying Eq. (12) to a series of compression tests on a sand-asphalt mixture produced the following expression [Vakily and Fitzgerald, 1972] where the constants were evaluated from constant strain rate "step" relaxation tests:

$$T(t) = 310\left\{\left(\frac{|\varepsilon|}{||\varepsilon||_9}\right)^{4.32} \varepsilon(t) + \left[1 - \frac{|\varepsilon|}{||\varepsilon||_\infty}\right]\int_0^t (t-\tau)^{-0.8}\,\dot{\varepsilon}(\tau)d\tau\right\} \quad (13)$$

The moduli for a strain of 0.37% and 0.5% is shown in Figs. 5 and 6.

The constant strain rate tests for two different rates are given in Fig. 7.

The ramp relaxation test results are given in Fig. 8.

Using the expression, Eq. (13), derived from the above data, predictions shown as open circles and experiments, shown as solid lines, were conducted for

(1) an interrupted ramp strain input at two different strain rates, shown in Figs. 9 and 10. A comparison of the above predictions with those of linear viscoelasticity is given in Fig. 11

(2) a single constant strain rate "sawtooth" strain, shown in Fig. 12

(3) a repeated constant strain rate test cycled between a set maximum strain and zero load, Fig. 13.

Again, it is clear that the sand-asphalt material exhibits a degree of permanent memory similar to the filled polyurethane propellant and the PBAN propellant. Figures 5 through 13 are from [Vakily and Fitzgerald, 1972].

An expression for the stress such as

$$T(t) = f[U(t), ||U(t)||_{L_\infty}] \quad (14)$$

produces a stress-strain relation with no relaxation but with the Mullins effect predominant [Mullins, 1947].

A mathematical justification for the above norms, and their restrictions, is given in [Hufferd and Fitzgerald, 1972] including thermodynamic implications.

In general, one may express the Cauchy stress tensor, T, as a function of the norms, for example

$$T = f[U(t),\ ||U||_{L_\infty},\ ||U||_{L_p}] \tag{15}$$

where U is the positive square root of the right Cauchy-Green (finite strain) tensor. The resulting polynomial function following the well known Rivlin-Spencer expression for initially isotropic materials will produce an expression up to the second power in the U and its norms with coefficients that are polynomials, or preferably here, rational fractions in the joint invariants

$$\mathrm{tr}U,\quad \mathrm{tr}U^2,\ - - ,\quad \mathrm{tr}||U||_{L_p},\quad \text{etc.}$$

Again, a three-dimensional expression similar to Farris' is derived for a single term, homogeneous form for T, namely

$$T(t) = A\left[\frac{\mathrm{tr}||U||_\infty}{\mathrm{tr}||U||_p}\right]^n (U(t) - 1) \tag{16}$$

Application of the above tensorial equation to a uniaxial test produce results similar to those previously shown.

2.0 AGING EFFECTS AND THE STEKLOV AVERAGE

The Lebesgue norms previously described are such that they produce the integral of the strain history on a P - summable basis. The use of a non-unit weighting function, $h_p(s)$, will provide for fading memory effects. The Lebesgue norms may have a basis in microscopic theory as presented by Farris in his doctoral thesis.

Nevertheless, the norms as used herein are simply continuum postulates whose use is justified by the results shown herein and in the various quoted references.

With the integrals as used, however, no aging effects are included.

Consider now a class of materials whose response is governed by certain weighted averages of the past strain history as well as by the present value of the strain and its several time derivatives.

A norm on such a space may be constructed as follows for a generalized input Λ

$$||\Lambda^t(s)||_{S_P^N} = \sum_{i=0}^{N} |\Lambda^{(i)}(o)|h_i(o) + \sum_{P=1}^{\infty} \left[\frac{1}{\text{mesD}} \int_D |\Lambda_r^t(s)|^P h_p^P(s)ds\right]^{1/P}$$

$$\text{with } \left[\frac{1}{\text{mesD}} \int_D h_p^P(s)ds\right]^{1/P} < \infty \text{ and } h_p^P(s) \geq 0 \quad .$$

and $\Lambda^{(i)}(o)$ the present i-th rates of change. (17)

The above is actually a semi-norm unless measD is finite.

Considering especially the second summand in Eq. (17), we shall call it a *Steklov Average* since it is a generalization of the Steklov-Lebesgue average given in [Kantorovich and Akilov, 1964].

A slight variation in the above, which will be termed the *modified Steklov Average* and which satisfies all the requirements of a norm (or semi-norm) is given by

$$||\Lambda||_{\overline{S}_P} = \left[\frac{1}{(1+k\ \mathrm{meas}D)}\int_D |\Lambda_r^t(s)|^P h_p^P(s)ds\right]^{1/P} \tag{18}$$

with $k \geq 0$.

Dropping the fading memory factor $h_p(s)$, for simplicity, results then in

$$||\Lambda||_{\overline{S}_P} = \left[\frac{1}{(1+kt)}\int_{s=0}^{t} |\Lambda|^P\ ds\right]^{1/P} \tag{19}$$

where we have taken $\mathrm{meas}D = t$ for the usual rather smooth physical inputs, Λ .

Looking at the Lebesgue norm ratios of Eq. (10) or Eq. (11) for example and the definitions Eq. (4) and Eq. (19) yields the following relation between the L_p norms and the S_p norms

$$\left[\frac{||\Lambda||_{\overline{S}_Q}}{||\Lambda||_{\overline{S}_P}}\right]^n = \left[\frac{||\Lambda||_{L_Q}}{||\Lambda||_{L_P}}\right]^n (1+kt)^{\left(\frac{Q-P}{QP}\right)n} \tag{20}$$

and for $Q \to \infty$,

$$\left[\frac{||\Lambda||_{\overline{S}_\infty}}{||\Lambda||_{\overline{S}_P}}\right]^n = \left[\frac{||\Lambda||_{L_\infty}}{||\Lambda||_{L_P}}\right]^n (1+kt)^{n/P} \tag{21}$$

It is readily shown that the ratio

$$\frac{||A||_{\bar{S}_Q}}{||A||_{\bar{S}_P}} \geq 1 \quad \forall\, Q > P, \quad P \geq 1 \tag{22}$$

since the dominance of L_Q over L_P is well known when $Q > P$ [Kantorovich and Akilov, 1964] and for $k \geq 0$, the multiplier is also ≥ 1 .

Defining T^L as the stress obtained from Lebesgue norms as in the previous section 1.0, the stress obtained by substituting modified Steklov norms is T^S where

$$T^S = T^L\,(1 + kt)^{\frac{Q-P}{QP}\,n} \tag{23}$$

or with $Q \to \infty$

$$T^S = T^L(1 + kt)^{n/P} \tag{24}$$

Consider for example a material governed by a simple permanent memory norm relation such as Eq. (6)

$$T_{11}{}^L = A\left[\frac{||\varepsilon_{11}||_\infty}{||\varepsilon_{11}||_{L_{21}}}\right]^n \varepsilon_{11}(t) \tag{25}$$

For a step relaxation test with strain magnitude ε_0 (25) yields

$$T_{11}{}^L = A\,\varepsilon_0\,t^{-n/P} \tag{26}$$

Applying instead the ratio of the modified Steklov norms S_P produces

$$T_{11}{}^S = A\,\varepsilon_0\,t^{-n/P}\,(1 + kt^*)^{n/P} \tag{27}$$

For a material which is strained immediately upon being formed, $t = t^*$. Otherwise however, t is the time relative to the beginning of the step strain whereas t^* is the actual time from the initial creation of the material to the present. The difference in behavior of the L_p and S_p norms is shown in Fig. 14. A step load-unload input is used with the L_1, $\bar{S}_1$ and $L_\infty = S_\infty = \varepsilon$ max. curves plotted for an example. After the load occurs, L_1 is constant as is L_∞. However, the S_1 norm reduces with time since it is averaging over the life of the specimen.

The relation between the L_p and S_p norms is, from (4) and (19)

$$||\Lambda||_{\bar{S}_p} = ||\Lambda||_{L_p} (1 + kt)^{-1/P} \tag{28}$$

It will also be noticed from Eq. (24) that the exponent of the "aging" term $1+kt^*$ is equal to the positive numerical slope of the inverse power law relaxation modulus, n/P .

Consider a typical solid propellant with n/P = 0.2 , then

$$T_{11} = A\, \varepsilon_0 t^{-0.2} (1 + kt^*)^{0.2} \quad . \tag{29}$$

If the relaxation modulus increases by 20% over a one-year ambient aging then $k \cong 3 \times 10^{-8}$, sec^{-1} .

A typical CTPB propellant increases its modulus by 20% in 100 days. With a slope n/P = 0.2 , hence, the value of k is 15×10^{-8} sec^{-1}.

It is also to be noted that for large values of kt^*, the aging is described by a straight line on a log-log scale. Further for n/P = 0, no relaxation and no strain rate effects, the aging is nonexistent.

Because of the small values of k, the use of the L_p rather than the S_p norms for short time loads on newly formed materials is fully equivalent. Further, for short loading times relative to the life of the material, $(1+kt^*)^{n/P}$ is essentially a constant and Steklov aging reduces to the so-called homothetic aging process [DeArriaga, 1969].

For finite strain, the essential results also hold. Consider an incompressible material subject to a simple step elongation with a stretch ratio of λ where λ is the stretched length divided by the initial length. Using a simple constitutive equation of the form of (16) but with modified Steklov norms instead of Lebesgue norms as in (21) results in

$$T(t) = A\left[\frac{\lambda+2}{\lambda+2\lambda^{-\frac{1}{2}}}\right]^n t^{-n/P} (1 + kt^*)^{n/P} (\lambda - 1) \tag{30}$$

For a material stretched when newly formed and then held at the stretch ratio, λ, with $t = t^*$

$$T(t) = A\left[\frac{\lambda+2}{\lambda+2\lambda^{-\frac{1}{2}}}\right]^n (t^{-1} + k)^{n/P} (\lambda - 1) \tag{31}$$

For long times then as $t \to \infty$,

$$T(t) = A\left[\frac{\lambda+2}{\lambda+2\lambda^{-\frac{1}{2}}}\right]^n k^{n/P} (\lambda - 1) \tag{32}$$

One could obviously extend the expansion to higher order terms in U, but the essential point to be made is that the material does not fully relax as $t \to \infty$. For the typical material previously mentioned, if the relaxation modulus at one second were 1000 psi, the final modulus would reduce to 60 psi. In addition, there can be considered an elastic component with no loss of generality.

3.0 CONCLUSIONS

It has been proposed that the characterization of relaxing, rate sensitive materials be based upon certain weighted averages of the past history which are semi-norms called modified Steklov averages.

The rationale is based upon previously mentioned guidance from molecular theory but is primarily based upon the postulate that the present response of a material is governed by its present deformation gradient and a selected weighted P-summable Steklov average (17).

Since in engineering practice, one seldom knows the detailed past of a structures thermal and deformation history but usually knows the average and the maximums, no serious loss of applicability should result.

The justification for the use of the proposed norms is based upon the several examples given herein where the predictions using norms is shown to be superior in accuracy to the predictions based upon linear viscoelasticity. The use of the $\overline{S}_\infty$ or L_∞ maximum value norms implies that the material also is sensitive to the maximum strain value it has ever been subjected to. The possible extension of the concept to viscoplastic materials is under study.

Farris has shown that for a selected class of solid propellants the use of the conventional time-temperature superposition integral is valid. The present author suggests that the time temperature shift integral

$$\xi = \int_{\tau=0}^{t} \frac{\tau}{a_T} \, d\tau \tag{33}$$

for reduced time ξ with the temperature shift factor a_T be used in the modified Steklov norm for aging effects. It is, of course, equally possible to use an absolute reaction rate correction for the kt term in (19).

The use of a rather simple homogeneous fraction involving only the max. norm and the P-norm of the modified Steklov average (25) produces three specific results for infinitesmal deformations

- .the relaxation modulus is described by an inverse power law, $t^{-n/P}$
- .the constant strain rate curves are described by a power law whose exponent is unity plus the relaxation modulus exponent, $t^{1-n/P}$
- .the relaxation and secant moduli are subject to an age hardening process n/P described by the factor (1+kt)
- .for very long term stretching and aging, the material stress response is nonvanishing as shown in (32).

It has been also shown that for short aging times the form of the equations reduces to ratios of Lebesque norms only. For short term loading relative to the age of the material, a homothetic aging process is produced.

There does not exist a unique inverse for strain as a function of stress, i.e., a general creep inverse. Indeed, an inverse does not generally exist unless a nonconstant weighting function, $h_p(s)$ is used. Even then, there results a nonlinear integral equation of the type

$$\phi(s)^m = \int |\phi(s)|\ h_p(s)ds\ ;\quad \phi(s) = \Lambda^P(s) \tag{34}$$

with the exponent m a rational fraction whose value will generally be near unity.

Since the present value of stress is governed by certain averages of the past strain history, uniqueness in the inverse is not to be expected. It is therefore suggested that a creep law be formulated in the same fashion as (25), for example

$$\varepsilon(t) = f[||T||_{\bar{S}_p},\ ||T||_{S_\infty}\ ,\ T(t)] \tag{35}$$

made specific as

$$\varepsilon(t) = B\left[\frac{||T||_{\bar{S}_P}}{||T||_{S_\infty}}\right]^m T(t) \tag{36}$$

which for a constant stress, T_0, results in

$$\varepsilon(t) = B\ T_0\ t^{m/P}\ (1 + kt)^{-m/P}\ . \tag{37}$$

The calculation time necessary to use the various norm equations given herein is much shorter for general inputs than the time needed for visco-elasticity. This computer time saving results from the fact that the norms are only a number at the present time whose value changes by the modified average at each step. If, however, nonconstant weighting functions, $h_p(s)$, are used, no computational advantage results.

4.0 ACKNOWLEDGEMENTS

The author wishes to acknowledge the past and continuing efforts of his colleagues R. Farris, W. Hufferd, J. Vakily, and M. Quinlan in pursuing and contributing to the ideas herein presented.

This work was sponsored in part by the Office of Naval Research under Contract No. 00014-67-A-0325-0001 and the Air Force Office of Scientific Research under Contract No. 72-2332.

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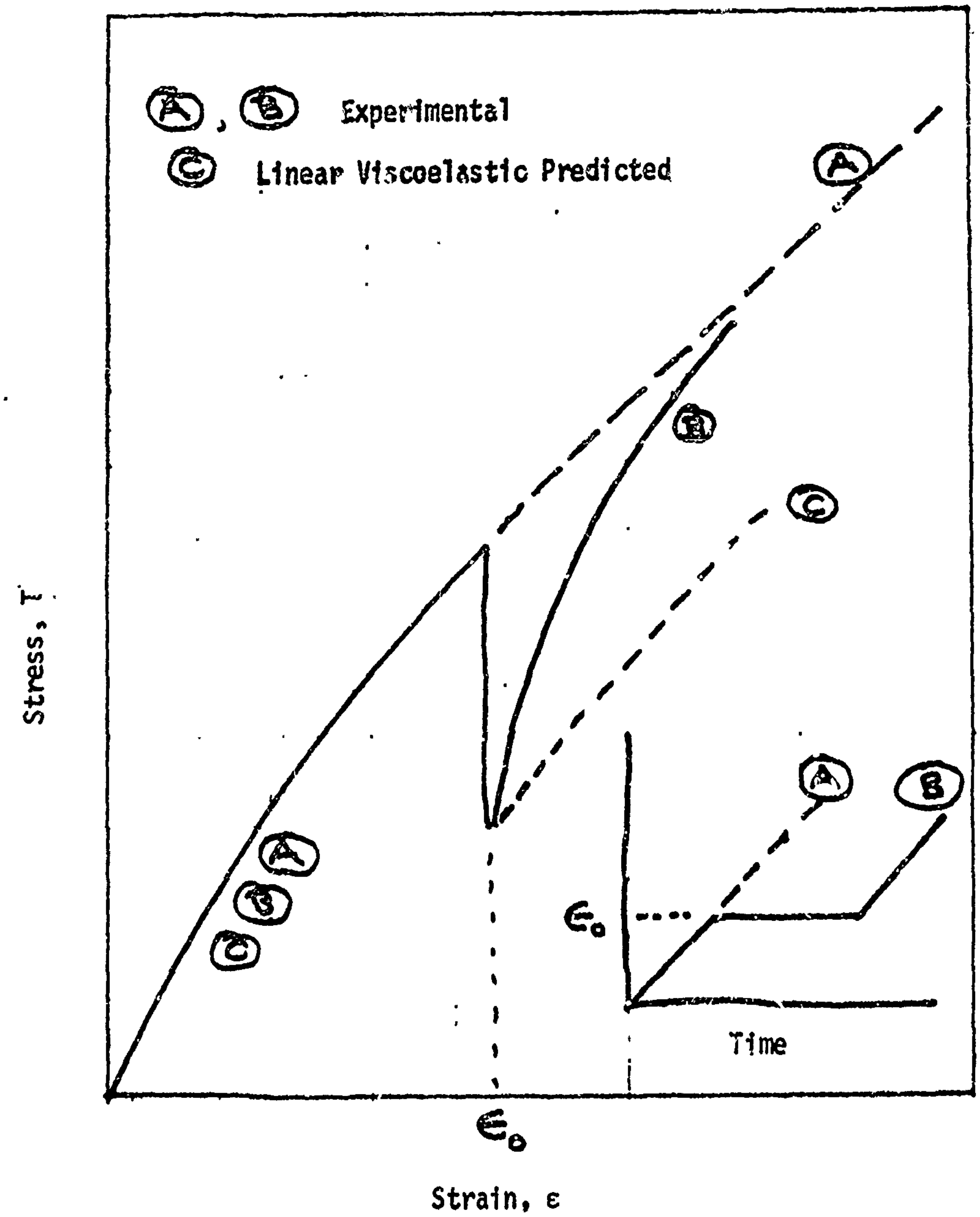

Figure 1 - STRESS FOR AN INTERRUPTED CONSTANT STRAIN RATE TEST

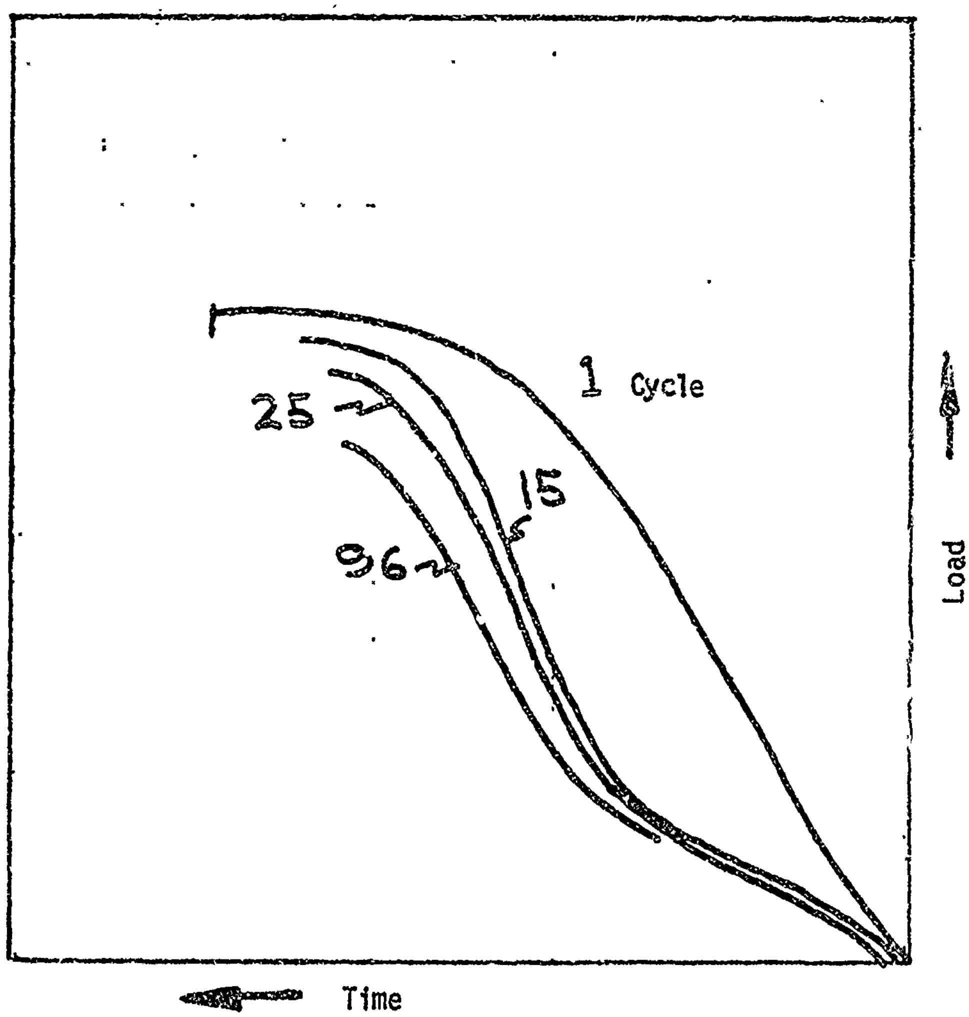

Figure 2 - UNIAXIAL CONSTANT RATE CYCLING OF PBAN PROPELLANT

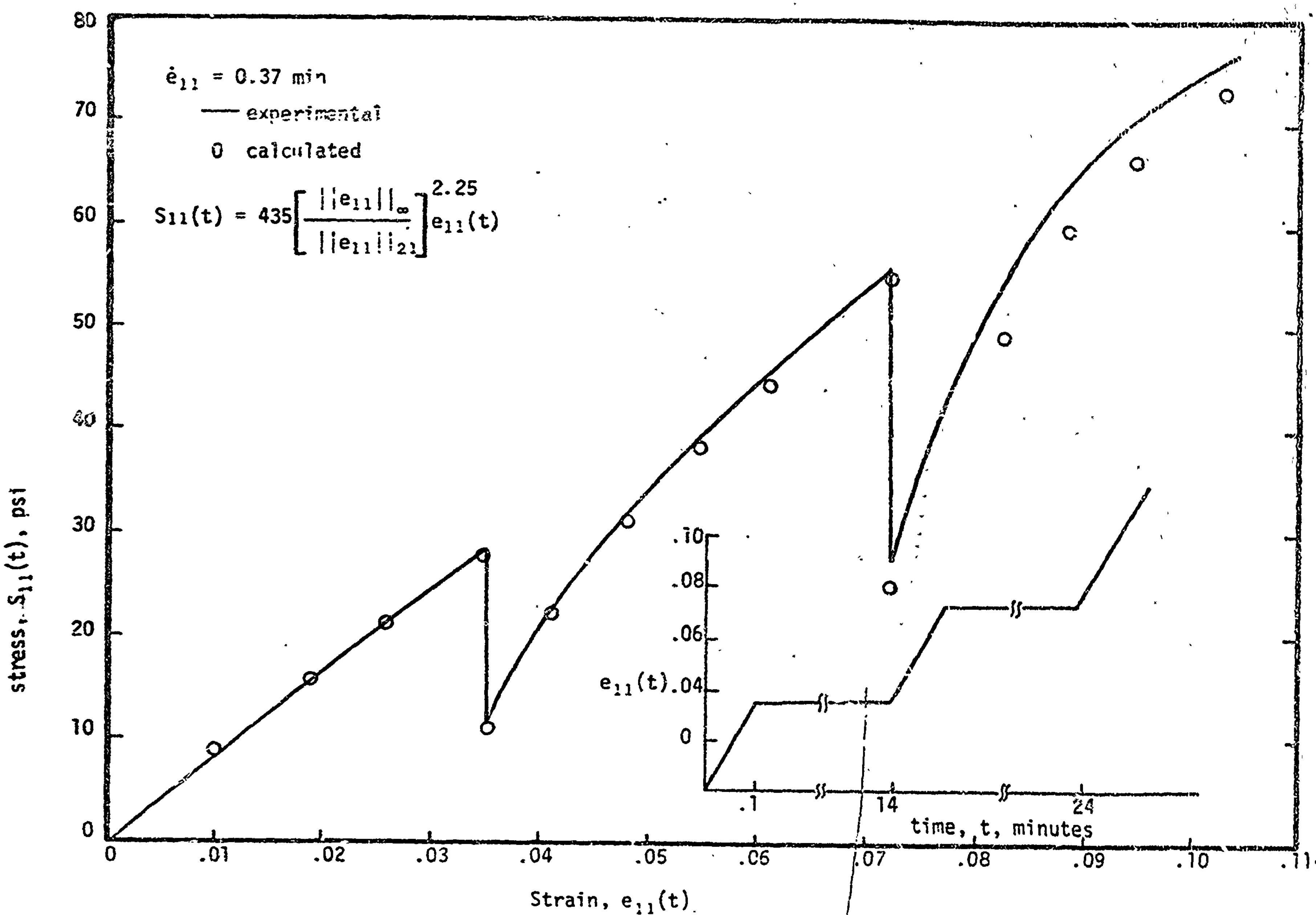

Figure 3 - COMPARISON OF CALCULATED AND OBSERVED STRESS-STRAIN OUTPUT FOR AN INTERRUPTED RAMP STRAIN INPUT

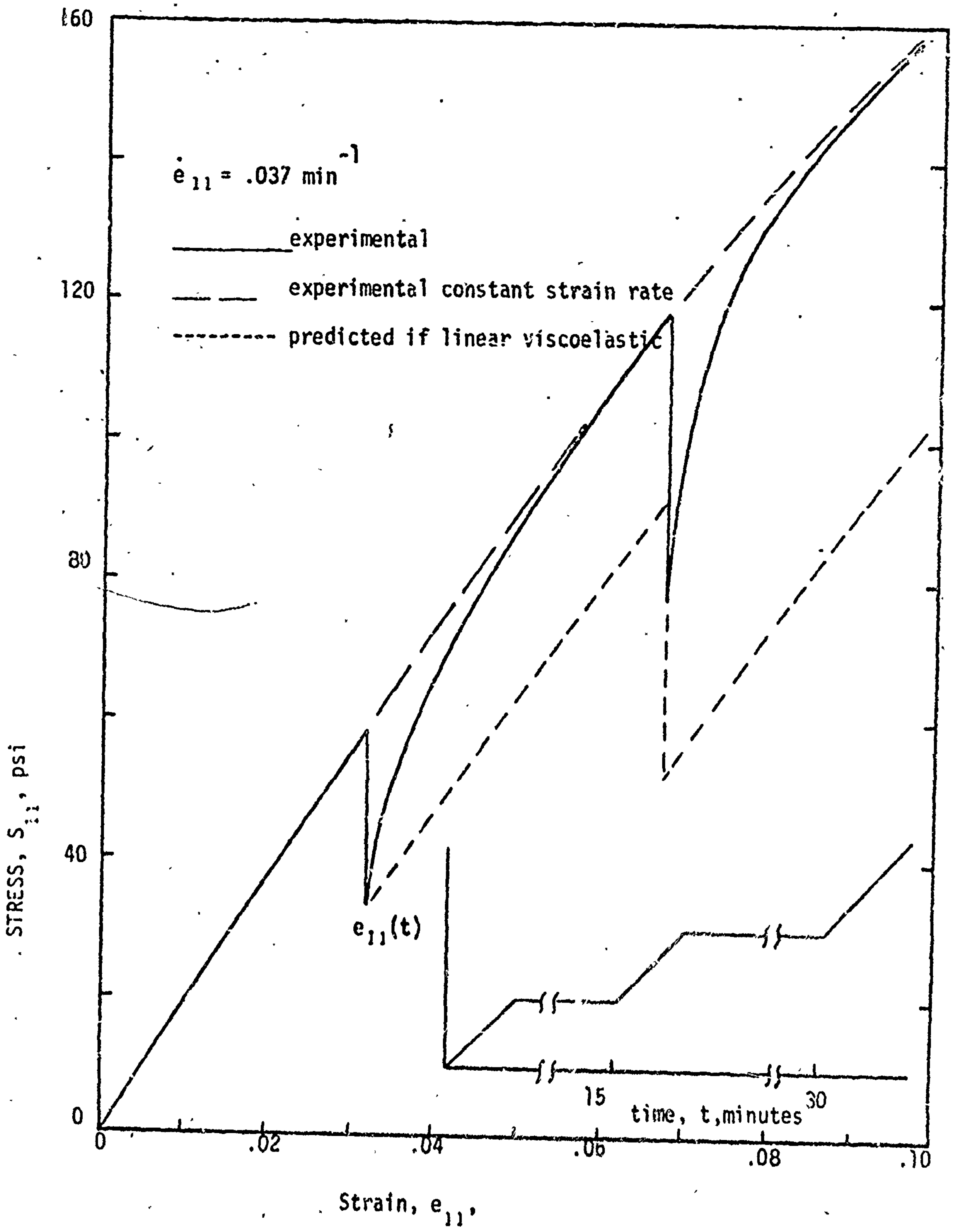

Figure 4 - STRESS OUTPUT FOR INTERRUPTED CONSTANT STRAIN RATE TEST

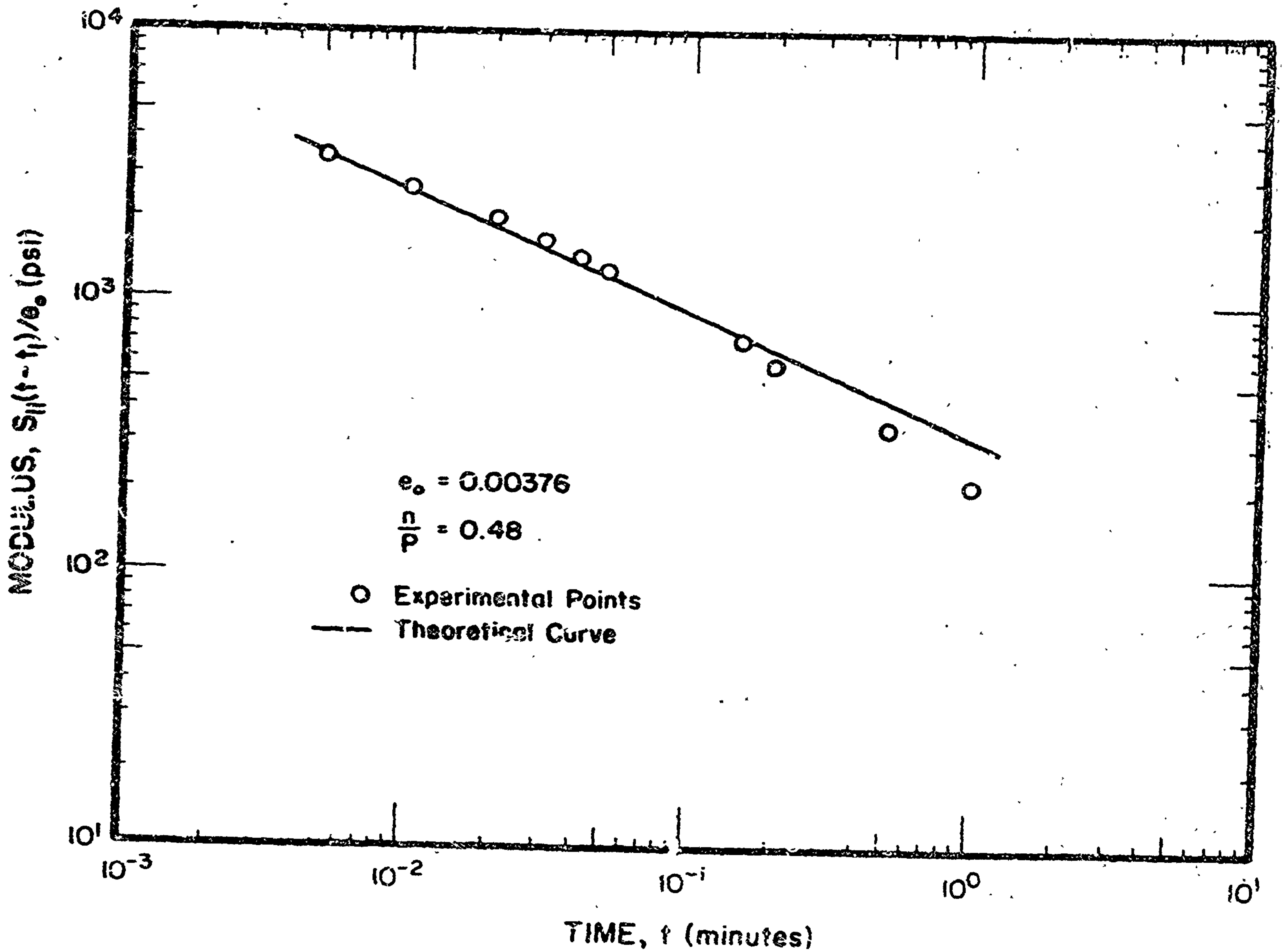

Figure 5 - RELAXATION MODULUS OF SAND-ASPHALT FOR FIRST TEST

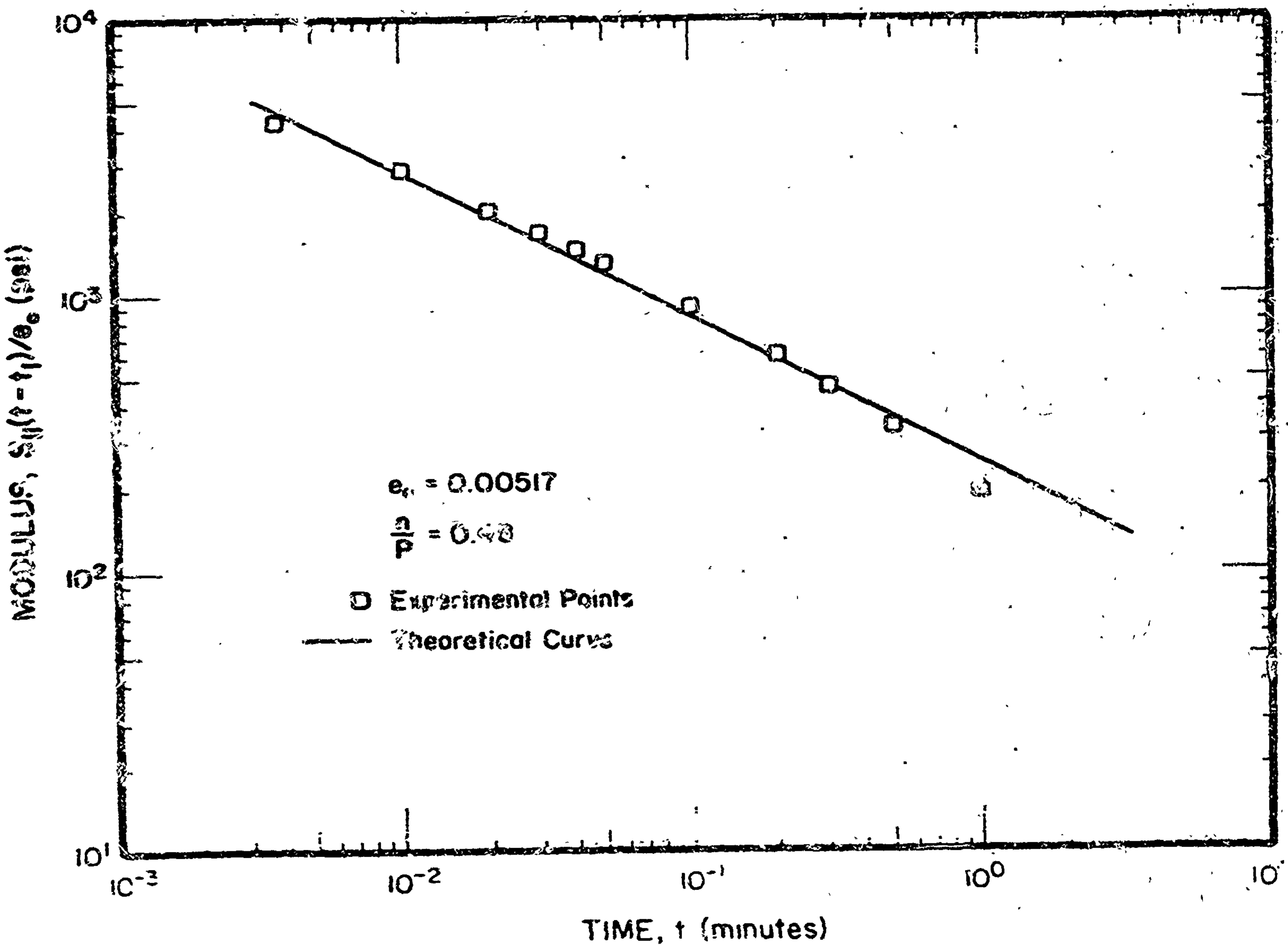

Figure 6 - RELAXATION MODULUS OF SAND-ASPHALT FOR SECOND TEST

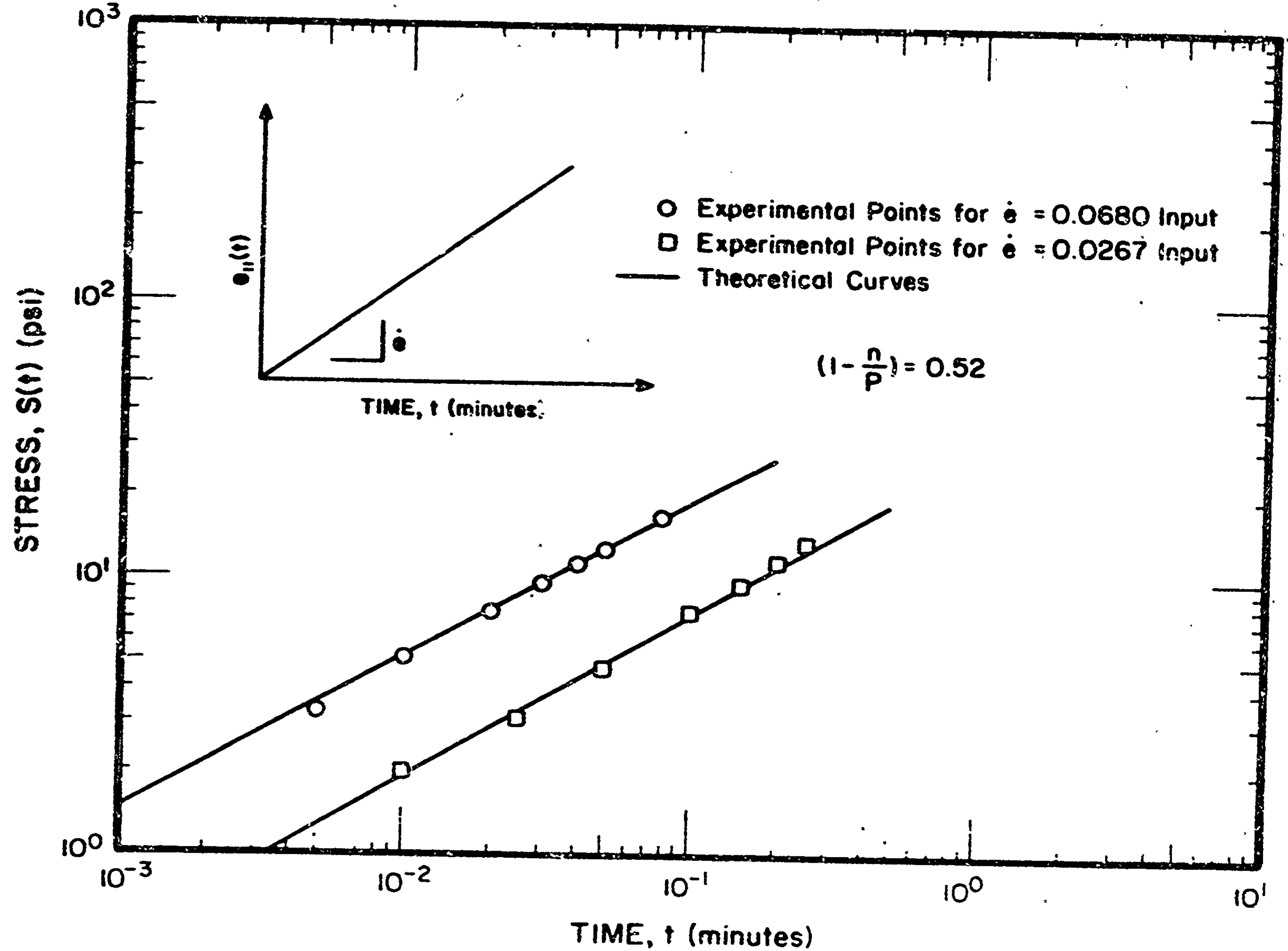

Figure 7 - STRESS OUTPUT FOR TWO DIFFERENT CONSTANT STRAIN RATE TESTS

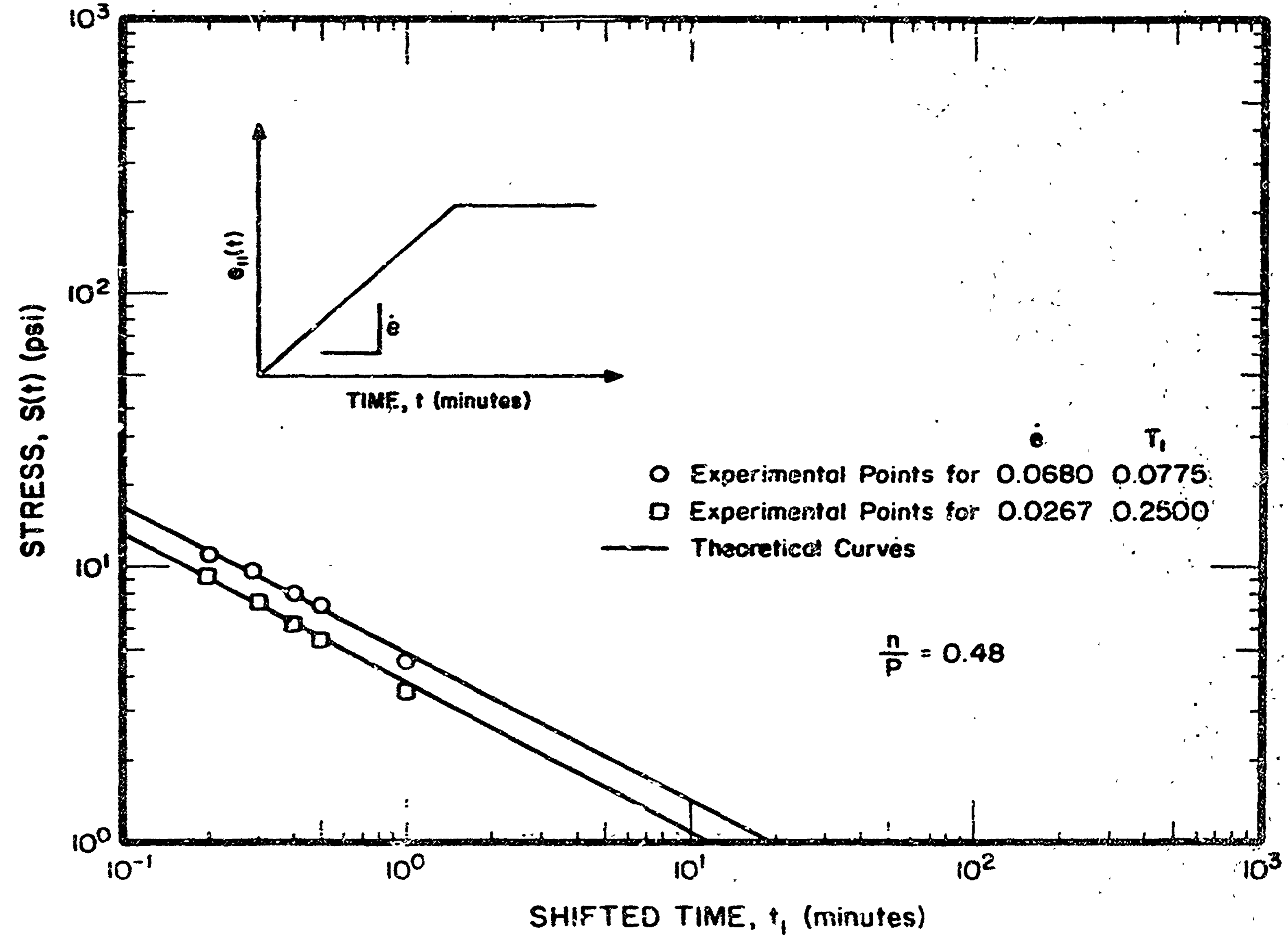

Figure 8 - STRESS OUTPUT FOR TWO DIFFERENT RAMP RELAXATION TESTS

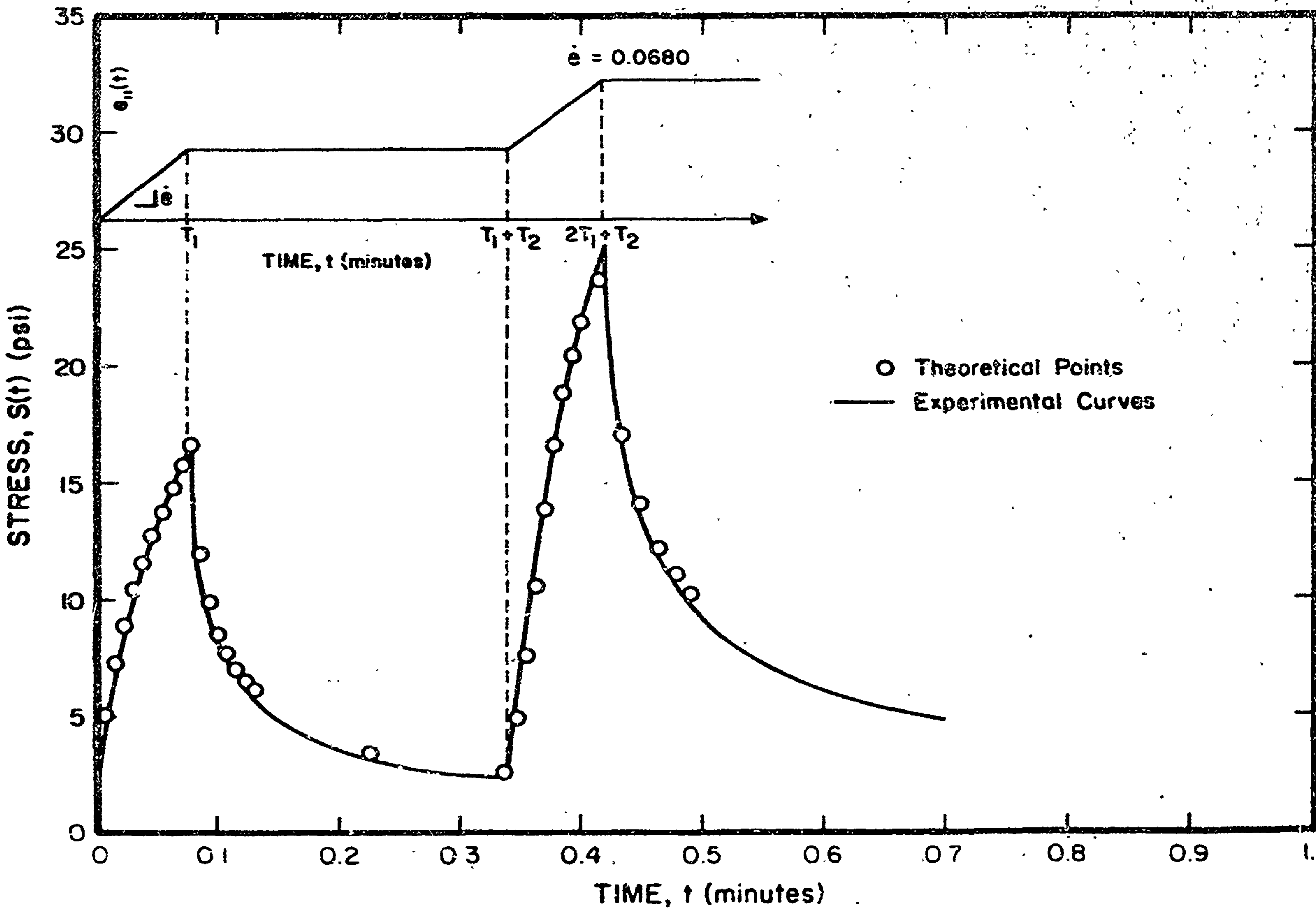

Figure 9 - STRESS OUTPUT FOR INTERRUPTED RAMP STRAIN INPUT

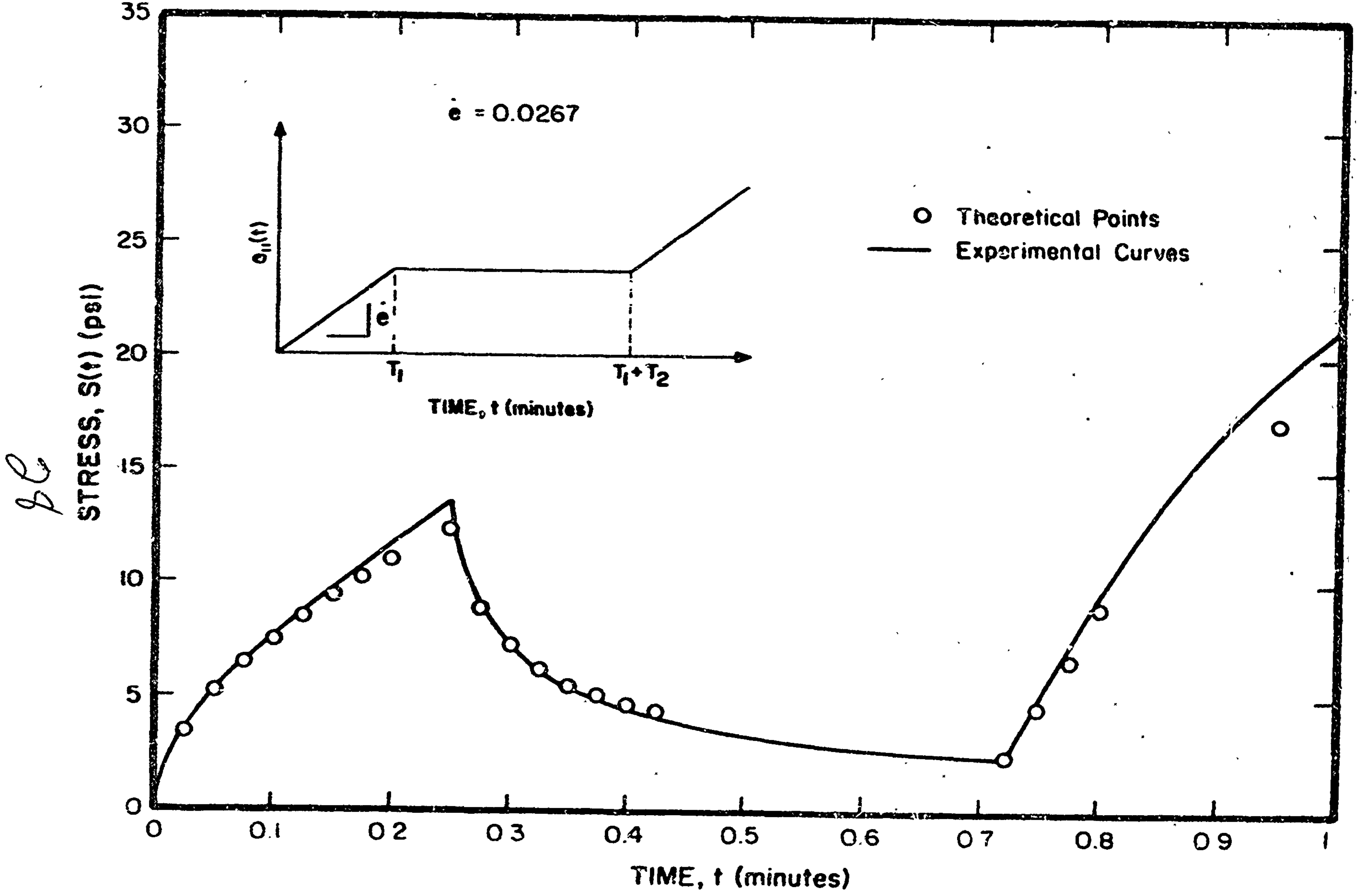

Figure 10 - STRESS OUTPUT FOR INTERRUPTED RAMP STRAIN INPUT

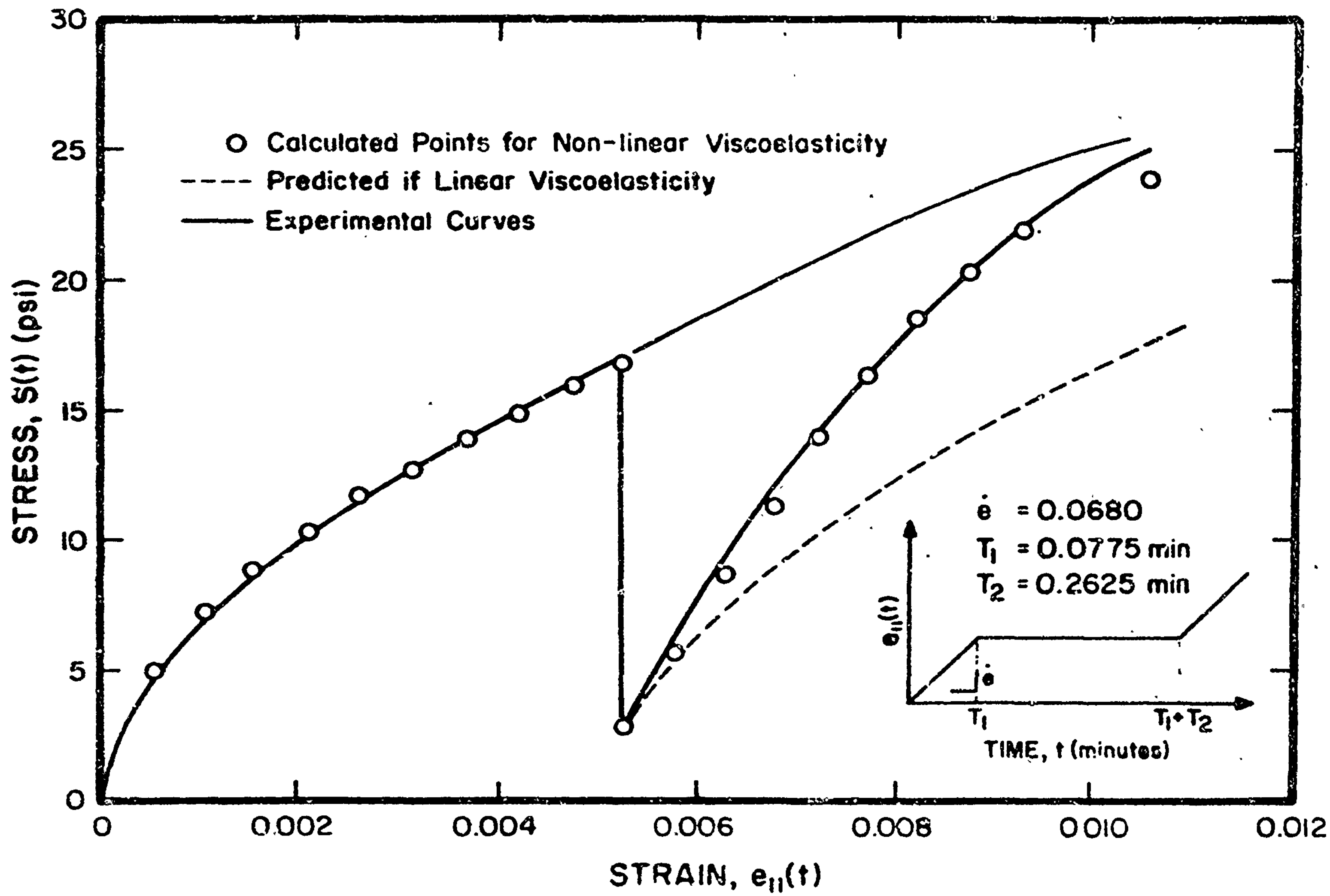

Figure 11 - COMPARISON OF CALCULATED AND OBSERVED STRESS-STRAIN OUTPUT FOR AN INTERRUPTED RAMP STRAIN INPUT

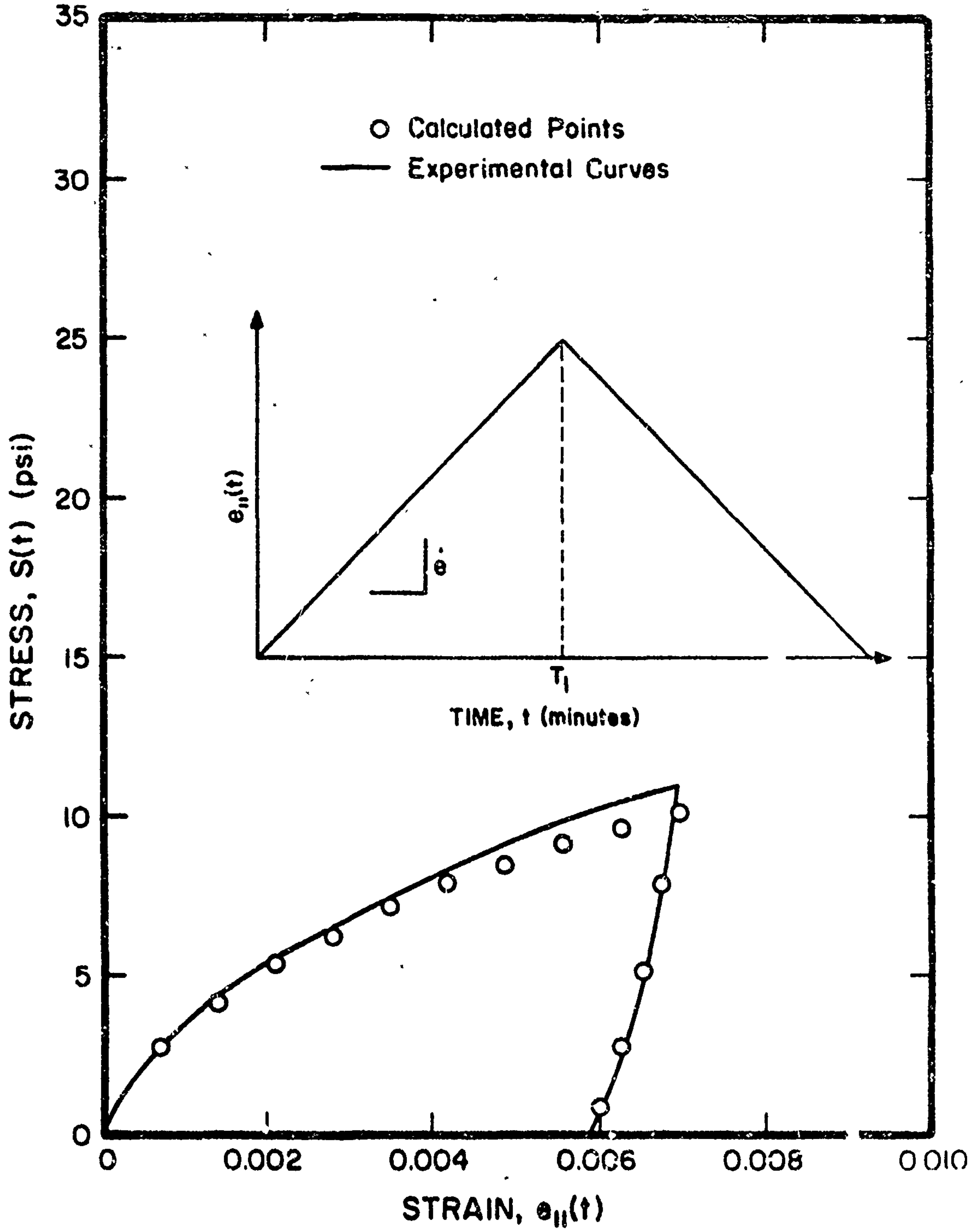

Figure 12 - COMPARISON OF CALCULATED AND OBSERVED STRESS-STRAIN BEHAVIOR OF SAND-ASPHALT

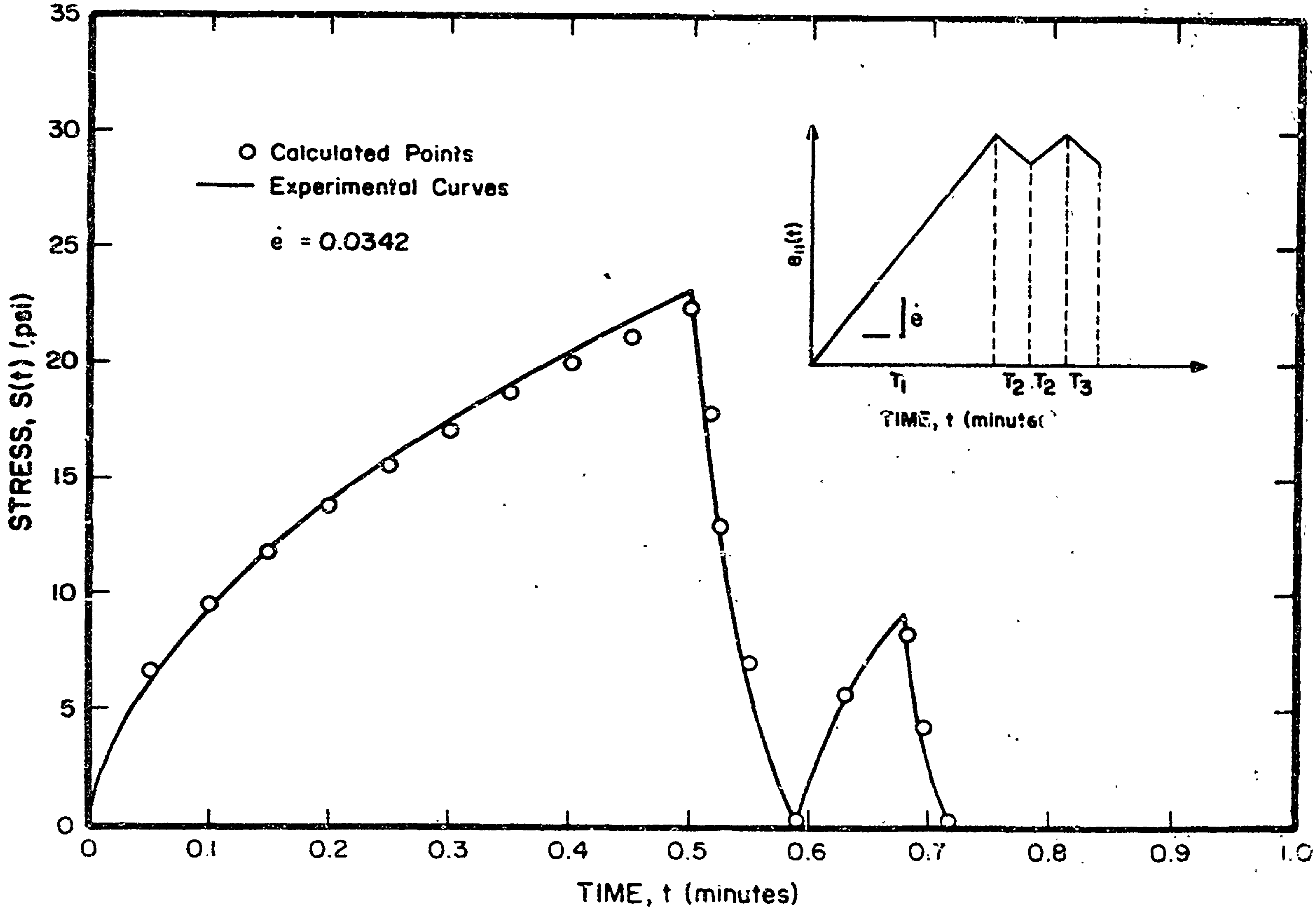

Figure 13 - COMPARISON OF CALCULATED AND OBSERVED STRESS OUTPUT FOR A REVERSED RAMP STRAIN TEST

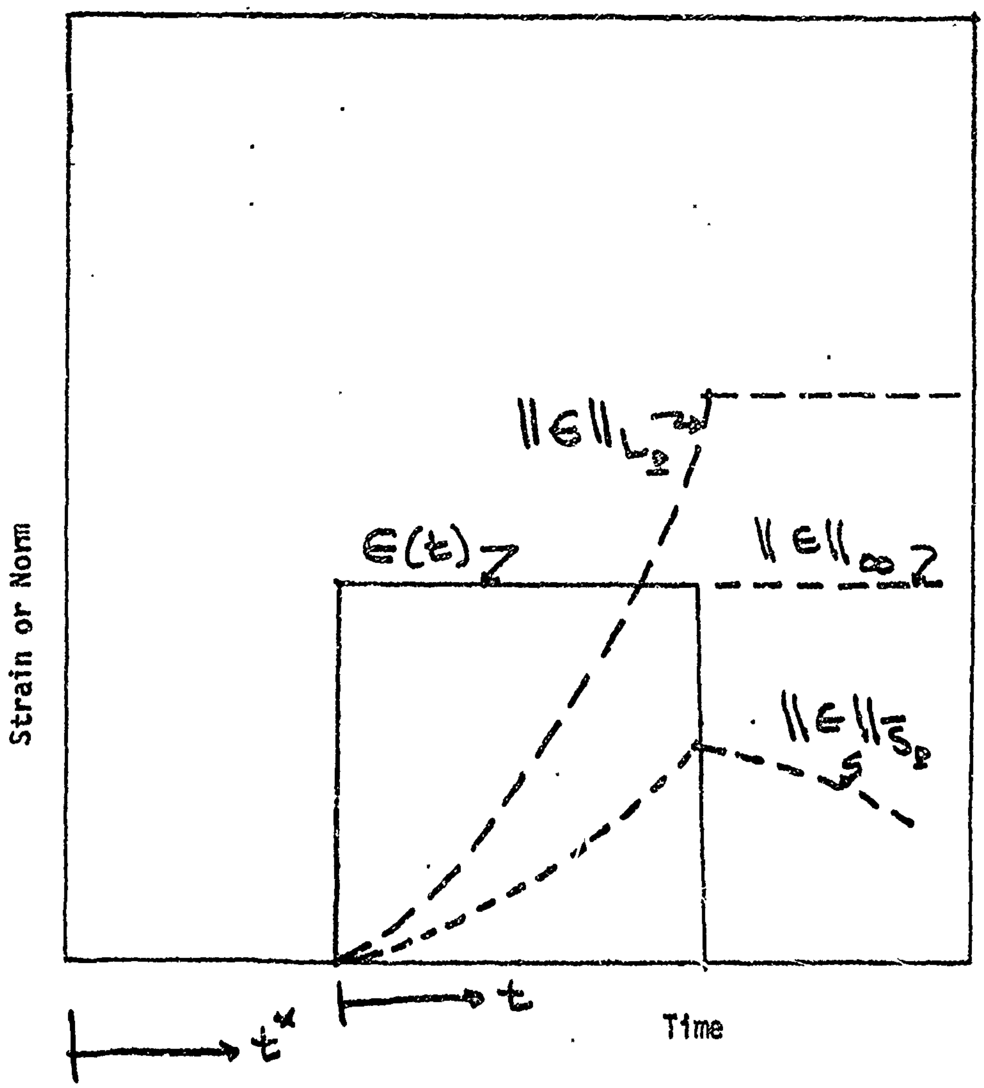

Figure 14 - TIME VS. NORMS